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Author: Parker

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H. M. Parker, Section Chief

REVIEW OF AIR MONITORING PROCEDURES AT CLINTON LABORATORIES

By H. M. Parker

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REVIEW OF AIR MONITORING PROCEDURES
at
CLINTON LABORATORIES

I. INTRODUCTION

The main emphasis on the measurement of radiation in air at Clinton Laboratories was initially placed on the measurement of activity on the plant site and environs due to the gases emitted by the separations plant stack. The function of these measurements was to be two-fold, (1) to establish that there was no current radiation hazard on or around the plant site from radioactive gases (2) by the correlation of radiation measurements with meteorological observations to arrive at evidence useful for the conditions at W. The extension of meteorological observations both here and at W during the past year has indicated that conditions are so variable that the probability of obtaining useful information from a comparison of the behavior of the discharged gases at the two sites is rather low. The radiation levels found here at Clinton are so low that it is difficult to conceive of conditions changing to an extent that would produce a serious radiation hazard. The function of the measurements is aimed, therefore, mainly at keeping records to indicate that the general population in and around the Plant site has not been exposed to any hazard from radioactive materials in the atmosphere.

It has been proved by calculations that the hazard from external total body radiation from a radioactive atmosphere is greater than the internal radiation from inhaled and absorbed material, with few exceptions. One special case has to be considered, namely that of radioactive Iodine, all of which will be readily absorbed by the body and about 20% of it retained by the thyroid gland. Continued exposure to radioactive Iodine at a concentration of 10^{-13} curie/cc has been calculated to be sufficient to deposit and keep deposited in the thyroid gland enough to cause 1 r exposure per day to that gland. The tolerable concentration for iodine if the limit were defined by external radiation would be 7×10^{-12} curie/cc. For Xenon, the tolerance concentration is taken as 2.3×10^{-11} curie/cc; for argon as 1.8×10^{-12} curie/cc.

If any appreciable portion of the activity in the air were due to Iodine, the interpretation of the external radiation measurements would be difficult. Evidence from the work on stack gas monitoring indicates however, that relatively little of the radioactive Iodine escapes from the stack. As a check on the possible accumulation of radioactive Iodine in personnel, selected members of the staff were tested for radiation activity of the thyroid gland. None was found.

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II. CONTINUOUS MEASUREMENT OF THE BETA AND GAMMA ACTIVITY IN THE AIR

Victoreen Integrons

The Clinton program called for the use of Victoreen integrons with recorders for measuring and recording the atmospheric radiation at selected points on and near the plant site and near K-25, Y-12, Oak Ridge Village and the Solway Gate. Special housings were built so that the atmospheric air would have free access to the ionization chambers but so that the parts containing the recorder and other mechanism would be kept warm. In this manner, atmospheric air containing beta and gamma emitters could come in contact with the integron chamber. The regular integron chambers are a bakelite composition material about 1.6 mm in thickness which will absorb nearly all beta rays with energy below .7 mev. No attempt has been made to date to install chambers on the integrons which will read low energy beta rays. This change has been omitted partly because (as shown below) some x-ray and gamma emitters are in air contaminants and would register, but mainly because we have been unable to bring the integron program into operation. Up to the time of this writing the Health-Physics Section has no useful records of activity as measured by an integron outside of the plant buildings or on the Reservation. Failure is due to late delivery of the integrons and to the failure of the integrons to operate satisfactorily after delivery. There is a general feeling that a record on the chart of a Leeds and Northrup recorded has more value than a series of entries in notebooks. I do not subscribe fully to this point of view. It should be emphasized that an L & N chart is useless for legal records, unless the measuring conditions that lead to the chart are (a) proper for the possible radiations, (b) clearly defined in the records. The overall picture can frequently be substantiated better by written notes of observations. Nevertheless, the relative ease of filing charts for future reference is appreciated. To this extent the Clinton program has been a disappointment.

III. INTERNAL MEASUREMENTS OF ACTIVITY IN THE AIR

X-22 Ionization Chambers

In the absence of the originally planned procedure, other methods of obtaining data on the activity of gas on the reservation have been used.

The X-22 chambers (Figure 1) are condenser chambers virtually of the type of blown-up pocket meters. The chamber length is about 8" and the outside diameter 7". The collecting volume is .4900 cm^3, and the electrostatic capacity is 10 cm.

The chambers have bakelite walls 1/8" thick, lined with aquadag. There are not metal parts exposed inside the chambers, and, therefore, they will be approximately wavelength dependent and suitable for radiation measurement down to rather low energy x-rays, but all except the very energetic betas will be absorbed in its walls. The chambers have been calibrated against radium gamma-radiation, but it is known that the wavelength error will not exceed 10% over the range contemplated.

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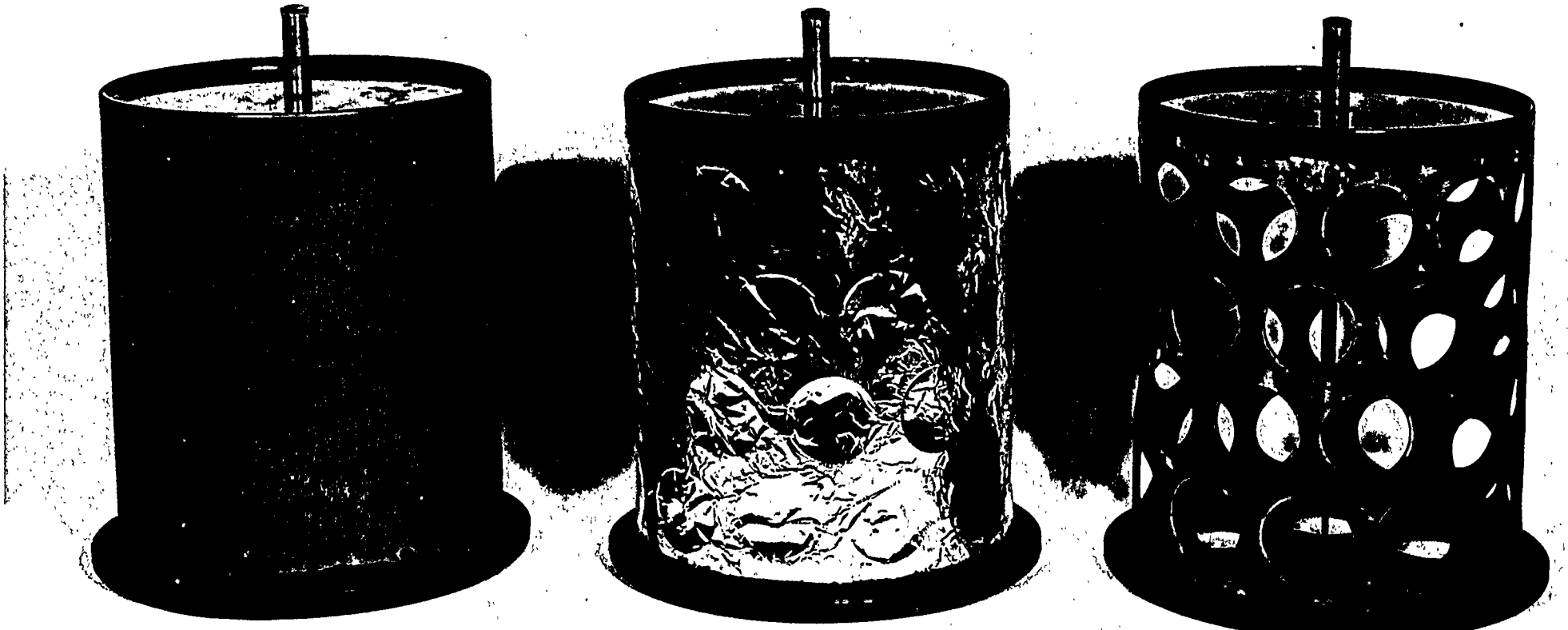

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To measure the discharge of these X-22 chambers in the field a regular Victoreen minometer was converted to battery operation for portable use. One scale division of the 0-100 mr scale of the minometer represents 4×10^{-13} milliroentgens exposure of the X-22 chamber. A plot of scale readings against exposure is linear over the entire scale and beyond. The scale was extended to cover more than the original 100 divisions by making additional marks on the scale. In this extended region, however, the voltage of the central electrode is low, being of the order of 20 volts and saturation conditions are somewhat open to question. At the low levels of intensity to which these chambers are subjected there is no evidence in practice of a failure of saturation.

Fifty percent of the chambers were converted so as to be sensitive to beta-rays by making 48 large holes over the surface of the bakelite wall. These holes were covered in some cases by cellophane with inner graphited surface, and in other cases by aluminum foil 0.001" thick. Strict insistence on wavelength independence leads one to require the cellophane type, but in practice the aluminum foil has been found to be much more durable. The X-22 meters were planned to be left outside in all weather condition, being made with drip-proof lips to prevent leakage of moisture into the chambers. The cellophane chambers were not weather proof, as the cellophane expanded, contracted and wrinkled with changes in weather conditions. Moreover, the violent rains occasionally directly punctured the upper cellophane wall. No such difficulty has been experienced with the aluminum wall chambers.

Considerable time was spent in preparing the insulators of the X-22 chambers in such a way that the leakage was satisfactorily low. The technique used was to cut the surface of the insulator clean and dry on the lathe and to polish the surface by lint-free paper with polystyrene dust. Under this treatment the insulation properties of the insulators increased for as much as one week after the operation. Insulators of proper quality were such that the leakage of the chamber was 3 or 4 divisions/hour. This corresponds to 0.012 to 0.016 mr/hr or to between 7 and 10 ion pairs/cc/sec. This is a very rational value of the natural background of residual ionization on the plant site. In other words, the actual insulator leakage accepted was between 0.1 and 1 div/hr. This corresponds to insulator resistance between 10^{19} and 10^{18} ohms. The insulator assembly was first tested in small chambers to remove the large leakage component due to actual ionization. Although insulators of this quality were laborious to prepare, there have been no subsequent difficulties either from humidity or other causes, despite the fact that these meters have been out in the field in all atmospheric conditions without the use of a desiccant in the chambers.

[redacted]

Daily measurements have been made at the following points; (indicated on the map, Figure 2), near the west rabbit cages on the plant site, the west hill #1, the west hill #2, the strawberry patch on the east side of the plant, the east valley, location A-8, A-12 and A-21. The average reading over a 24-hour period has at no time exceeded three times the natural background at the given location. This background is the limiting value of accuracy of the method.

J. S. Cheka has followed the correlation between wind direction and stack discharge and the X-22 readings. Whenever the wind direction was favorable for the passage of stack gas over a measuring station, a reading significantly in excess of the natural background was obtained. The effect was independent of whether the 205 stack was discharging active gas or not. This apparent paradox led to the conclusion that there was another source of active gas in approximately the same location.

A review of the situation indicated, of course, that this source was the activated argon emerging from the 105 stack. The estimated quantities of active gas emitted at the present time are as follows:

	Average curies/day	Relative Ionization γ	$\beta+\gamma$
105 Stack-Argon	200	740	1500
205 stack - Xenon	11	1.2	5.3
205 Stack - Iodine	1	1	1.6

Not only is the amount of argon greater than that of the others, but its more energetic radiation emphasizes the effect in the X-22 chambers. Since the high readings over a 24-hour period average not more than 0.024 mr/hr above background the iodine contribution must be about 3×10^{-5} mr/hr or a concentration of 5×10^{-17} curie/cc. This is well below tolerance. The Xenon contribution does not exceed 10^{-4} mr/hr or a concentration of 5.5×10^{-16} curie/cc. Let it be conservatively assumed that the present average iodine concentration is 10^{-15} curie/cc. Then, if the argon hazard were eliminated, the Clinton power could be increased by a factor of 100 without exceeding tolerance for iodine or xenon in the stack gases. This is an encouraging situation with respect to W forecasts.

Under adverse wind conditions, the stack gases are brought to ground on the site for periods up to 2 minutes. 1 mr/hr has been so observed and this might be due mainly to xenon and iodine activities. Such an event gives 0.033 mr, or 3.3 mr at 100-fold concentration. Of this, perhaps 1 mr could be due to iodine radiation. The corresponding amount of iodine would be below the permissible daily amount for transfer to the thyroid. A major increase in Clinton power could, therefore, be tolerated even under adverse weather conditions.

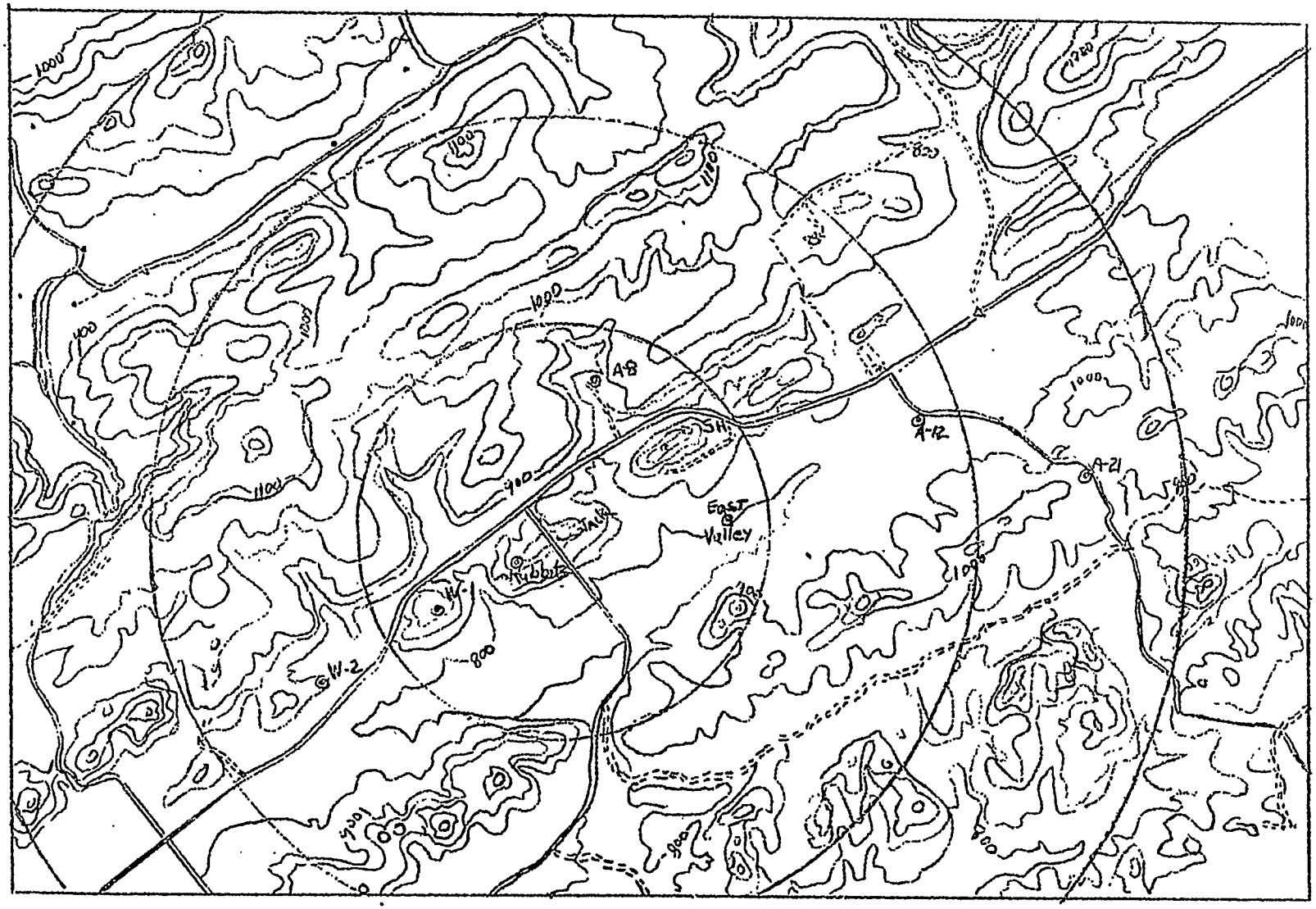

FIG 2. LOCATION OF STATIONS AROUND THE STACK. CIRCLES AT ½, 1 AND 1½ MILE RADII

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IV. NATURE OF THE INCIDENT RADIATIONS

Consistently higher readings are obtained with the beta plus gamma ray meters than with the gamma ray meters, in the ratio of 10 to 9. As the beta ray windows of the chamber occupy 42% of the wall, a complete beta ray wall would lead to a ratio of 1.25 to 1. This ratio seems to be the same whether there is activity above the natural background or not; therefore, there seems to be no spectacular case of a strong beta emitter being present without accompanying gamma radiation in approximately normal proportion. The chambers have been exposed uniformly on special stands at a height of 5' above ground. This cuts out the effect of any local irregularities in the ground level. It does make a slight uncertainty in the collecting volume for different radiations. Effectively, hard gamma rays will have been counted from a hemisphere, whereas soft radiation, either beta or x-ray will have entered from almost a sphere of radiation. Careful attention to this point would be required if the meters had ever indicated a significant level of radiation.

V. FILM METERS

Local monitoring with the X-22 chambers has been done on the basis of two or three circuits of the inner area per day. This would be a time-consuming procedure for the measurement of meters at the gates or the reservation or in the populated areas. Therefore, in the absence of the integron recorders planned for the gates on the reservation, x-ray film has been placed in these locations and metered on a basis of once per week. This has been done routinely since 4/13/44 at the Eliza Gate, Solway Gate, White Wing Gate, and the Oliver Springs Gate. The regular film badges were used for this purpose and both beta and gamma ray measurements were obtained. There has never been a measurable record in a one-week period. In the interest of Security, the film badges were contained in long wooden boxes with open louvred sides to allow entrance of air containing possible soft beta components. The boxes also contained a recording maximum and minimum thermometer and a special evaporation meter. These were installed primarily so that if the boxes were opened by unauthorized persons the main interest would not be drawn to the film badges. It was planned that one of the films would be left in a given location for a period of one month to see if there were accumulated exposures measurable in that time, but owing to the method of interchaning the boxes so that they are not opened in the presence of guards or other spectators this has not been done. Some film measurements have also been made of the area at Lenoir City by special arrangements and these, too, have at no time shown a measurable activity.

VI. SPOT CHECKS OF AREAS SUSPECTED OF TEMPORARILY INCREASED ACTIVITY

During periods of discharge from the Separations plant stack it is possible to follow the course of the activity by observing the brown nitrous fumes. In the absence of evidence to the contrary it is generally

[redacted]


DO NOT DISTURB
20
10
0
10
20
30
40
50
60
70
80
90
100
110
120

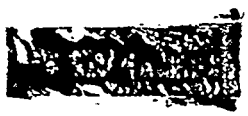
assumed that the active elements will follow the path pursued by the brown fumes. In this manner, it is possible to obtain spot checks on the radiation at ground level by pursuing the plume of brown fumes, under favorable wind conditions. This has been done somewhat sporadically both by means of ionization chambers and counters. A thin-walled Lauritsen electroscope has been used in general for this work. There are now eight entries in the record of the brown fumes descending to the ground in concentrations detectable by sense of smell. On three of these occasions a measurable radiation level was found. Within the confines of the plant, levels of 1 mr/hr for a period of 1 or 2 minutes have been observed. Further afield where the smoke is liable to touch the ground for a longer period but after greater dilution, no measurable activity has been recorded to date. It is possible though that had counters been used at the right time some further readings would have been obtained. It is planned by K. Z. Morgan to install, in the integron houses, counters that will operate in this way. These would give spot indications of short-term increases in the atmospheric activity. These readings will be of particular value for correlation with the meteorological conditions, but as far as any actual hazard to personnel is concerned, the method of integrating the dosage over an 8-hour or 24-hour period is entirely adequate.

VII. LABORATORY METHODS OF GAS MONITORING

Two methods of measuring the concentration of active gas in and around the laboratory have been used.

1. Open Counters

Portable glass-walled counters with a scaling circuit or occasionally with a counting rate meter have been used to measure activity. With the equipment available here it has been found practicable whenever possible to use AC operated equipment in preference to the available battery models. K. Z. Morgan has designed several models in which different caps can be placed readily over the counter to sample beta rays of different energy and to give some measure of gamma activity. With reference to the calibration, it is assumed that any beta particle will produce about 75 ion pairs per cc. in air, from which the conversion of beta counts to ionization in terms of reps (roentgen equivalent physical) can be made. If N be the number of counts per minute in a thin-walled counter of flat plate area A cm^2 the dosage rate is approximately $\frac{2.2 \times 10^{-3} \times N}{A}$ mrep/hr. Gamma ray calibration



can best be done by exposure of the tube to a radium source. The use of counters for health measurements of quantum radiation is valid only for high-energy radiation.

The method has two objections, namely (1) the results are disturbed by the presence of beta contamination on surfaces in the vicinity, (2) it is not possible to follow the decay of the gaseous activity due to the transient nature of the conditions when gases are being emitted.

2. Standard Gas Sampling

Sampling has been carried out by strong steel cans of volume approximately 10.5 liters, which can be evacuated and taken to the measuring sites. When a gas sample is required, the Hoke valve on the can is opened to admit gas to the vessel. In this manner, either a series of samples can be taken simultaneously at various locations, or a successive series at different times in the same location. Balance of the gas monitoring equipment consists of a counter in a specially constructed lead chamber which is air tight. This chamber is previously evacuated, being sure that the high voltage is removed from the counter before the evacuation proceeds. Active gas is then shared between the sampling can and the evacuated chamber. The volume of the counting chamber is approximately 1.7 liters; the final pressure after sharing gas, is 0.86 atmosphere. The vacuum requirements of a closed chamber are, therefore, not stringent, and it is possible to maintain the sample in the chamber for several days with little loss of gas.

The method used today has been semi-quantitative only. For accurate measurements, it would be necessary to standardize the system by samples of known concentrations of the required active gases. It has been sufficient to extrapolate the count obtained in the small counter chamber to that which would be obtained at the center of an infinitely large sphere of the same nature of active material, by applying an extrapolation factor. For beta radiation, this factor is approximately the ratio of the range of the particles of average energy to the effective radius of the counting chamber. For high energy particles (for example the beta particles of radioactive argon) this factor is approximately 60, a close acceptance of the numerical value would be out of order. For gamma rays in the gas, the extrapolation method is no more satisfactory. Elements measured to date have been argon, xenon, iodine. The half-life of radioactive argon A^{41} as measured by the system is 117 minutes. There is no ready explanation for the excess of this over the accepted 110 minutes, as the obvious correction factors would both tend

to reduce the apparent period. If the counter chamber leaks, then either air will enter and increase the absorption of radiation in the gas, thereby reducing the count in the chamber, or the radioactive gas itself will escape from the chamber. In either case, the half-life would decrease. For the activity of the 5.4 day xenon, the extrapolation factor is only 3 and, therefore, the results are more nearly quantitative. On the one occasion when a strong concentration of radioactive iodine was sampled, the method broke down. It was found that the counting rate was unchanged when the chamber was evacuated and refilled with clean air. Investigation indicated a layer of radioactive iodine deposited on the counter tube and on the walls of the vessel. This experiment was fruitful only in verifying the 8-day half-life of the iodine. Presumably, a large fraction of the activity had also settled out on the inside of the sampling can before the test was completed. Procedure was evidently quite unsuited for the sampling of the vapor.

3. Measurement of Alpha Activity

Alpha particle activity in the air has been investigated mainly by the precipitator method. Air was drawn through a precipitator of the Cottrell kind, modified to have a flow of 11-1/2 cu.ft. of air/minute instead of the 3 cu.ft./minute of the commercial model. A metal tube was substituted for the original glass tube of commercial models and this was lined with aluminum foil on which the dust and active particles were precipitated by electrostatic action. The aluminum foil was transferred to a special linear-amplifier chamber for direct alpha count. Attempts to assay the concentration of long-lived alpha emitters such as the dust from uranium and thorium metals were obscured by the deposition of active deposits of radon and thoron existing naturally in the atmosphere. The precipitator method was used to determine the concentration of radon and thoron in the air as a quantitative check on the method. The observed concentrations of radon have ranged from 10^{-17} curie/cc to 5×10^{-16} curie/cc and for thoron from 8×10^{-19} curie/cc to 10^{-17} curie/cc*. These can be compared with the accepted values for radon concentration ranging from 2×10^{-17} to 10^{-15} curie/cc. The close agreement of the precipitator results with those obtained by standard methods indicates that the precipitator method is not far from quantitative. The collection efficiency of the precipitator is not accurately known, nor is it certain that it is always constant. The value used has been 70% efficiency at a flow of 11-1/2 cu.ft./minute.

* Radon concentration is greatly affected by rain, etc. At a given location it is sensitive to changes originating elsewhere, since the 3.85-day half-life of radon gives time for considerable travel through the air.

[redacted]

Almost 100% efficiency has been demonstrated at a flow of 3 cu.ft./minute, and whenever the activity to be checked is sufficiently high it is good practice to use the precipitator at a reduced rate of flow. The concentrations of radon or thoron in the air are obtained by reference to the special decay curve of the activity over the given period of observation. These have been evaluated by Coveyou. After 6 hours, the radium active deposit of short-life has virtually disappeared and the residual activity is then made up of the thorium active deposit plus long-life emitters. The decay of the thorium active deposit is governed at all times by the period of Th B(10.6 hours) and this half-life is troublesome for the accurate measurement of low activities. If a count is determined 6 hours after the collection of samples and again on the following day, the fraction of the total count due to the long-life activity can be computed. If P be the number of counts/min in the special cylindrical chamber at an estimated 45% geometry, and the air flow was 11.5 cu.ft./minute for a time T minutes, then the concentration M of the long-life emitter is -

$$M = 4.4 \times 10^{-18} \times \frac{P}{T} \text{ curie/cc, or}$$

$$4.4 \times 10^{-15} \times \frac{P}{T} \text{ microcuries/liter.}$$

The activity can readily be translated into micrograms/cc when the nature of the emitting substance is known. Further details of the precipitron procedure as used here and some measuring results of the activity in the laboratory will be given elsewhere (CN-H 1892)

J. S. Cheka and T. S. Block have made the observations with the X-22 chambers. D. J. Rendell and T. W. Bloss have done most of the measurements with the precipitrons. L. H. Weeks has operated the film badges at the gates. Other monitoring devices have been used by almost all men in the Section.

*NOTE; Slight editorial changes made by Dr. R. S. Stone and not reviewed by H. M. Parker.

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